CHARACTERIZATION OF LOW LEVEL RADIOACTIVE WASTE FROM THE AGS COMPLEX INCLUDING AN ASSESSMENT OF PURE BETA (β) EMITTERS

Introduction

The following is a characterization of low level radioactive waste from the AGS complex including an assessment of radionuclides that are exclusive beta (β) emitters.

Summary

Measurements made over the last few years at AGS indicate the activity per unit mass of waste ranges from 1 to 5 nCi/g. About 2000 to 3000 cubic feet of low-level radioactive waste is generated each year. After several months of decay, which occurs before the waste is packaged and shipped off-site, the predominant measured nuclide is 5.271 year ^{60}Co. AGS waste is segregated into bins according type of material; for example, iron, aluminum, copper. The radionuclide ^{60}Co is present in each major category of the AGS segregated waste stream. A comparison of cross section, half-life and abundance of target nuclides relative to ^{60}Co yielded a first-order estimate of activity per unit mass of waste for pure β^- emitters. Accordingly, ^{3}H and ^{63}Ni may be expected at 1 nCi/g or less, and ^{14}C at 50 pCi/g or less. The nuclides ^{59}Ni, ^{93}Mo and ^{151}Sm were also considered similar to pure β emitters since they would be difficult to detect via their low-energy photon emissions. Accordingly, ^{59}Ni may be expected at 1 pCi/g or less, ^{93}Mo at 0.4 pCi/g or less, and ^{151}Sm at 10 aCi/g or less.

The Alternating Gradient Synchrotron and Radioactive Waste

The Alternating Gradient Synchrotron has been in operation for 32 years providing protons for the high-energy physics program, and in addition for the past nine years providing heavy ions for the nuclear physics program. At this time, the AGS has embarked on an ambitious high-energy physics program to look for physics beyond the Standard Model. In the area of nuclear physics, the heavy-ion physics program has entered a virgin physics domain and is in the process of characterizing the nature of the interactions and searching for evidence of the quark-gluon plasma and the production of strangeness enriched matter. This part of the experimental program is the natural lead in to the next generation of experiments to be performed at the Relativistic Heavy Ion Collider starting at the end of this decade.

Most if not all radioactive waste at AGS is due to residual radioactivity created by the accelerated proton beams when they are stopped in accelerator materials. The total accelerated proton beam (H^-) allowed at the Linac is $9x10^{17}$ GeV-nucleons per hour. Most of this beam is used after it reaches 200 MeV in energy and most goes to a BNL Medical Department facility for the production of radiochemicals, and their refinement into radiopharmaceuticals. The Medical Department waste stream is separate from the waste generated at the AGS complex. Only a small percentage of the total Linac beam is sent to the AGS Booster. At the Booster, two electrons are stripped off and the beam is

accelerated to about 1.5 GeV. It is then transported to the AGS where further acceleration begins. The total accelerated proton beam (H^+) output from AGS is about $6x10^{13}$ protons per spill for AGS experimental areas.

Since the AGS can accelerate all ion species, a generic measure of other accelerated ion species is stated in terms of nucleons. For example, Au ions have 197 nucleons. For ion species other than H^+, AGS accelerates about 10^{11} nucleons per spill. The ions reaching the AGS are originally accelerated at the Tandem van De Graaff facility where they reach several hundred MeV per ion. These heavy ions are injected into the AGS Booster. Spills of ions or protons from the AGS to the experimental areas are at the frequency of once every 2 to 3 seconds.

The energy of the accelerated protons may be as high as 30 GeV, but generally they are 24 GeV. The heavy ion energy is about 10 GeV per nucleon. Most accelerated ions end up at the targets inside heavily shielded target halls that typically have 12-foot thick high-density-concrete walls. During transport through many hundreds of yards of transport lines, and many hundreds of yards of accelerators, a small percentage of the ion beam is lost. The accelerators are typically shielded with 20 foot roofs and 60 foot sides of earth berm. The lost beam interacts along beam-line in items such as magnets, concrete and earth shielding, vacuum pipes and cables. Uncollided accelerated protons that end in the target halls are stopped in iron beam dumps that are up to 50 yards long. The target halls, or target caves as they are sometimes called, tend to be the locations where the highest level of residual radioactivity is measured. Target caves contain items similar to items found in transport lines, plus they contain small targets which are usually a few ounces of platinum. About half of the beam may interact only once in the target. The other half proceeds to the iron dump, and secondary particles emitted from the small target interact in nearby magnets and concrete shield walls.

Most of the residual activity in beam line components is due to the long term operation of the high-intensity proton program which typically runs for 20 weeks per year, and has run for the past thirty years. Most items in the complex are re-used year after year. However, the AGS produces about 2000 to 3000 cubic feet of uncompressed low-level radioactive waste each year.

Activation of Materials At AGS

Stopping high-energy particles will initiate sub-microscopic reactions in the nuclei of atoms of beam pipes, magnets, cables, shields, targets and beam stops. These initial interactions produce secondary particles such as neutrons, other protons and pions. The pions behave like protons, only they are less massive and are short lived. The nuclei of atoms struck by these high-energy particles will fragment. This fragmentation results in a range of lower mass nuclei and many lower energy particles like neutrons. Tens of neutrons 'boiling-off' per interaction of a high-energy particle on a nucleus is not uncommon. The fragments of the struck nucleus are radioactive. The fragments produced are proton-rich and hence decay by positron emission (β^+) or electron capture

(EC). Most are short lived, minutes to days. Positron decay produces 0.511 MeV annihilation electromagnetic radiation that is easily seen via standard gamma spectroscopy. In effect, there is no such thing as a pure β^+ emitter. EC produces many x rays since the electron shell vacancies fill up again. This is also observed via standard spectroscopy. Some of the lower energy neutrons that are 'boiled-off' from the initial interactions of the high energy particles are absorbed by the other nuclei, thus increasing the mass of the struck nucleus and making it unstable or radioactive. These neutron-rich nuclides decay by β^- emission which is often accompanied by high-energy photon (MeV) emission from the decay of short-lived metastable states of the daughter nuclide. The most common of these neutron-rich nuclides is ^{60}Co. It is largely produced from the n,γ capture reaction on ^{59}Co. For the most part, very few pure β^- emitting nuclides are formed; that is, very few β^- emitters decay directly to the ground state of a stable daughter nuclide. However, three pure β^- emitters are may occur in the AGS accelerator environment and they are: ^{3}H, ^{14}C, and ^{63}Ni.

The materials used in construction of the AGS experimental areas are limited in number, the most important being iron, steel, copper, aluminum, concrete, oil and plastic. These materials are generally not used in a pure isotopic form. That is, they have dissimilar metals at welds or braze, or they are alloyed with other metals such as iron, nickel or cobalt, or they consist of naturally occurring stable isotopes of the same element, or they are parts of complex beam-line components. Thus, irradiation produces a variety of radionuclides in any given accelerator item.

The AGS segregates its radioactive waste into different radioactive waste containers on the basis of the item's predominant composition. The categories are plastic, oil, concrete, aluminum, iron or steel, and copper. In the analyses of these categories of items, the AGS measures naturally occurring radioactive nuclides at their naturally occurring levels. These nuclides are ^{40}K, ^{214}Bi, ^{214}Pb, ^{212}Pb, ^{226}Ra, and ^{228}Th. The man-made β,γ emitting nuclides range in half life from days to years, and consist of:

Irradiated Material	Nuclides That May be Identified Via Their Photon Emissions
Plastic, Oil	^{7}Be, ^{22}Na, ^{46}Sc, ^{54}Mn, ^{57}Co, ^{60}Co, ^{68}Ga, ^{88}Zr, ^{113}Sn, ^{124}Sb, ^{125}Sb, ^{133}Ba, ^{134}Cs, ^{207}Bi
Concrete	^{7}Be, ^{22}Na, ^{46}Sc, ^{54}Mn, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{110m}Ag, ^{134}Cs
Aluminum	^{7}Be, ^{22}Na, ^{54}Mn, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{68}Ga, ^{95m}Nb, ^{110m}Ag, ^{133}Ba, ^{134}Cs
Iron, Steel	^{7}Be, ^{22}Na, ^{26}Al, ^{46}Sc, ^{54}Mn, ^{59}Fe, ^{56}Co, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{67}Cu, ^{68}Ga, ^{75}Se, ^{88}Zr, ^{88}Y, ^{95m}Nb, ^{97m}Nb, ^{101}Rh, ^{102}Rh, ^{110m}Ag, ^{113}Sn, ^{124}Sb, ^{125}Sb, ^{133}Ba, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{203}Hg, ^{207}Bi
Copper	^{7}Be, ^{22}Na, ^{54}Mn, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{68}Ga, ^{110m}Ag, ^{133}Ba, ^{134}Cs

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^{60}Co. As can be seen from above, ^{60}Co is present in each major category of the AGS segregated waste stream. Thus, comparison of cross section, half-life and abundance of target nuclides relative to Co-60 will yield an estimate of activity per unit mass of waste for pure β^- emitters.

Estimate of Pure Beta Emitters In AGS Waste

As indicated previously, several different pure β^- emitting nuclides are possible: 12.33 y ^{3}H (β^-), 5745 y ^{14}C (β^-), and 100 y ^{63}Ni (β^-). The nuclides 75,000 y ^{59}Ni (EC, ~0% β^+), 3000 y ^{93}Mo (EC), and 90 y ^{151}Sm (β^-, 0.9% 0.02 MeV γ) are also worth considering since they would be difficult to detect via their low-energy photon emissions if they are integrated into accelerator components. In the following table, the neutron capture cross-section for a specified target mass number is reported. The integral neutron cross section is reported; that is, thermal to 20 MeV neutron interaction. This is the most likely production method for these nuclides. At energies higher than 20 MeV, the total interaction cross-section approaches the geometric cross-section for the nucleus which is of the order of 500 mb for all target nuclides, plus the fluence of higher energy particles is much less for everything except the platinum targets.

Nuclide Production Cross Section Taken From BNL 17100, mb, Per Neutron Capture Interaction in Specified Target
(Item That Most Likely Contains Target Atoms Of Interest)

Nuclide of Interest	Natural Carbon (Plastic, Wood, Earth)	Oxygen-16 (Concrete)	Aluminum - 27 (Beam Pipes, Concrete)	Natural Calcium (Concrete)	Natural Iron (Magnets, Beam Dumps, Heavy Concrete)	Cobalt-59 (Magnet Steel, Earth, Concrete)	Natural Nickel (Stainless Steel, Copper Pipe, Nickel Alloys for Brazing)	Natural Copper (Magnets, Cables)	Natural Molybdenum (Steel, Nickel Alloys, Earth, Concrete)	Natural Europium (Earth, Concrete)
12.33 y ^{3}H		27 (n,t)	2.2 (n,t)	2.2 (n,t)	4 (n,t)	0.7 (n,t)				6.2 (n,t)
5745 y ^{14}C	1.5 (n,γ)	965 (n,p)								
75,000 y ^{59}Ni							83 (n,2n) 2250 (n,γ)			
5.271 y ^{60}Co						77 (n,γ)	665 (n,p)	27 (n, α)		
100 y ^{63}Ni							2250 (n,γ)	1.4 (n, He^3,β^-)		
3000 y ^{93}Mo									29330 (n,γ)	
90 y ^{151}Sm										2.1 (n,t) 17.1 (n,p)

For proton capture reactions, the following formula is taken from Barbier:

$$\sigma(Z_i, A_i) = f_1(A_T) f_2(E) \frac{P e^{-P(A_T - A_i)}}{1 - 0.3/PA_T} e^{-R\left(\left|Z_i - SA_i + TA_i^2\right|\right)^{3/2}}$$

where:

σ is the cross section for production of nuclide Z_i, A_i, millibarns,
f_1 is taken from Figure II.3 (see footnote 1),
A_T is the mass number of the target atom,
f_2 is 1, taken from Figure II.4 (see footnote 1),
P is 0.056, taken from Figure II.1 (see footnote 1),
R is taken from Figure II.2 (see footnote 1),
S is 0.486, and
T is 0.00038.

From this formula, a table may be constructed for proton capture. The nuclear reactions for proton capture considered were p,α, p,pn, p,γ, p,t and p,n on a target nucleus that could be present at AGS. Only p,pn reactions were found to produce the radionuclides of interest.

Nuclide Production Cross Section, mb, Per Proton Capture Interaction in Specified Target

Nuclide of Interest	Cobalt-59 (Magnet Steel, Earth, Concrete)	Natural Nickel (Stainless Steel, Copper Pipe, Nickel Alloys for Brazing)	Natural Molybdenum (Steel, Nickel Alloys, Earth, Concrete)	Natural Samarium (Magnets)
75,000 y ^{59}Ni	17 (p,pn)			
100 y ^{63}Ni		5.11 (p,pn)		
3000 y ^{93}Mo			65 (p,pn)	
90 y ^{151}Sm				0.41 (p,pn)

The materials present in the AGS waste stream vary in abundance. To first order samarium, molybdenum and europium are found in trace amounts, less than 10^{-3}, relative to cobalt, nickel and copper that yield ^{60}Co. Carbon, oxygen, and calcium are likely to be roughly equivalent to amount of cobalt, nickel and copper in the waste stream. Aluminum and iron are more abundant than cobalt , nickel and copper in the waste stream by a factor of about 10.

On the basis of cross section, half life and approximate abundance of target nuclides in the waste stream, the following table was constructed.

[1]Barbier, M., Induced Radioactivity, North Holland Publishing Company, Amsterdam, 1969.

Nuclide of Interest	Decay Factor Relative to ^{60}Co (A)	Production Cross Section Relative to ^{60}Co (B)	Abundance of Target Atoms Relative to Target Atoms Yielding ^{60}Co (C)	Estimate of Relative Activity to ^{60}Co (A x B x C)
12.33 y ^{3}H	0.43	0.055	10	0.2
5745 y ^{14}C	0.00092	1.3	1	0.01
75,000 y ^{59}Ni	0.000070	3.0	1	0.0002
100 y ^{63}Ni	0.053	3.0	1	0.2
3000 y ^{93}Mo	0.0018	38	0.001	0.00007
90 y ^{151}Sm	0.059	0.02	0.001	0.000001

Thus, ^{3}H and ^{63}Ni may be expected at 1 nCi/g or less, ^{14}C at 50 pCi/g or less, ^{59}Ni at 1 pCi/g or less, ^{93}Mo at 0.4 pCi/g or less, and ^{151}Sm at 10 aCi/g or less.